Appendix E

Source Terms

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DR WENZEL CONSULTING INC
Radiological Consequence Analysis
1560 Mountain Rose
Idaho Falls, Idaho 83402

November 3, 2004

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Subject: Recalculation of H-3 and I-129 activities for INTEC site CPP-31

Dear Lorie:

Current information indicates that the H-3 and I-129 activities calculated in Wenzel 1997 for INTEC site CPP-31 are high. Based on the following discussion, it is recommended that both the H-3 activity and the I-129 activity reported in Wenzel 1997 be reduced by a factor of 20.

The activity leaked to the soil in site CPP-31 was from the 2nd and 3rd cycle process wastes being transferred from deep tank WM-181. Some of the waste in WM-181 had been concentrated in the PEW evaporator; however, waste that exceeded the PEW evaporator limits was sent directly to WM-181. It is known that most of the H-3 remains in the evaporator overheads rather than the bottoms.

The current evaluation of the amount of H-3 leaked to the soil in 1972 is based on a sample of tank WM-181 in 1971 (Rhodes 1972). The activity ratio of H-3/Cs-137 measured in the WM-181 sample is 1.25E+03/8.81E+06 = 1.42E-04. The sample of WM-181was taken in September 1971 while the activity leaked to the soil was sampled in November 1972. Thus the activity in WM-181 had decayed for a period of approximately 14 months prior to leaking to the soil. The fraction of H-3 remaining after 14 months of decay is 0.9363. The fraction of Cs-137 remaining is 0.9735. Applying these decay factors to the H-3/Cs-137 activity ratio measured in 1971, the activity ratio calculated for 1972 is 1.42E-04 (.9363/.9735) = 1.36E-4.

The activity ratio of H-3/Cs-137 reported in Wenzel 1997 is 2.54E+04/8.96E+06= 2.83E-03. Dividing this ratio by that calculated above, 2.83E-03/1.42E-04, a reduction factor for H-3 of 20 was calculated.

Like H-3, most of the I-129 also remains in the PEW overheads. Based on Schlinder 1988, the maximum reduction factor for I-129 in the PEW evaporator is 100. However, wastes bypassing the PEW would have a much lower I-129 reduction factor. Unfortunately, no analysis of the I-129 in WM-181 was made. An observation made by Mike Swenson is that the activities of H-3 and I-129 in the PEW overheads tend to track one another. This suggests that the reduction factor for I-129 may be as low as a factor of 20. A reduction factor of 20 for the I-129 is therefore recommended.

The tables in Wenzel 1997 reported the released activity in D/s/mL and normalized to a total of 1 Ci of activity. Neither of these units of activity can be used in the current studies without conversion. The converted data normalized to 1 Ci of Cs-137 with corrections for H-3 and I-129 is presented in Table 1.

Table 1: Calculated 1972 Radionuclide Activities in Released Liquid for Site CPP-31

Nuclide	Half-Life		Activity Normalized to Cs-137
H 3	1.228E+01	yr	1.4E-04
Be 10	1.600E+06	yr	3.8E-11
C 14	5.730E+03	yr	1.5E-09
Si 32	4.500E+02	yr	2.3E-13
Cl 36	3.010E+05	yr	6.2E-13
V 50	4.000E+16	yr	1.3E-20
Co 60	5.271E+00	yr	1.1E-03
Ni 63	1.001E+02	yr	5.7E-14
Se 79	6.500E+04	yr	5.5E-06
Rb 87	4.730E+10	yr	3.7E-10
Sr 90	2.912E+01	yr	9.5E-01
Zr 93	1.530E+06	yr	2.8E-05
Nb 93m	1.360E+01	yr	1.3E-05
Nb 94	2.030E+04	yr	2.9E-10
Tc 98	4.200E+06	yr	5.8E-12
Tc 99	2.130E+05	yr	1.9E-04
Rh102	2.900E+00	yr	1.2E-08
Pd107	6.500E+06	yr	2.0E-07
Ag108m	1.270E+02	yr	1.6E-12
Cd113m	1.360E+01	yr	9.1E-05
In115	5.100E+15	yr	1.9E-16
Sn121m	5.000E+01	yr	9.8E-07
Te123	1.000E+13	yr	1.5E-19
Sb125	2.770E+00	yr	4.4E-03
Sn126	1.000E+05	yr	4.9E-06
I129	1.570E+07	yr	1.5E-08
Cs134	2.062E+00	yr	3.5E-03
Cs135	2.300E+06	yr	1.2E-06
Cs137	3.000E+01	yr	1.0E+00
La138	1.350E+11	yr	2.7E-15
Ce142	1.050E+11	yr	3.8E-10
Nd144	2.100E+15	yr	1.8E-14
Sm146	1.030E+08	yr	8.2E-14
Pm147	2.623E+00	yr	2.2E-01
Sm147	1.070E+11	yr	1.3E-10
Sm148	8.000E+15	yr	3.3E-17
Sm149	1.000E+15	yr	1.7E-16
Eu150	3.600E+01	yr	9.5E-12
Sm151	9.000E+01	yr	1.2E-02
Eu152	1.360E+01	yr	7.1E-06
Gd152	1.080E+14	yr	8.1E-19
Eu154	8.600E+00	yr	4.3E-03
Eu155	4.960E+00	yr	5.1E-03
Ho166m	1.200E+03	yr	9.0E-11
TM171	1.920E+00	yr	3.0E-15
Pb210	2.226E+01	yr	7.1E-12
Ra226	1.600E+03	yr	4.9E-11
Ra228	5.750E+00	yr	2.2E-15
Ac227	2.177E+01	yr	3.1E-10
Th228	1.913E+00	yr	1.3E-08
Th229	7.340E+03	yr	7.3E-13

Table 1: Calculated 1972 Radionuclide Activities in Released Liquid for Site CPP-31 (Continued)

Nuclide	Half-Life		Activity Normalized to Cs-137
Th230	7.700E+04	yr	1.6E-08
Th232	1.405E+10	yr	4.6E-15
Pa231	3.726E+04	yr	1.5E-09
U232	7.200E+01	yr	2.3E-10
U233	1.592E+05	yr	1.1E-10
U234	2.445E+05	yr	2.6E-06
U235	7.038E+08	yr	9.4E-08
U236	2.342E+07	yr	1.4E-08
U238	4.470E+09	yr	2.2E-08
Np236	1.150E+05	yr	2.5E-12
Np237	2.140E+06	yr	1.5E-06
Pu236	2.851E+00	yr	2.6E-09
Pu238	8.775E+01	yr	4.6E-04
Pu239	2.413E+04	yr	2.6E-04
Pu240	6.569E+03	yr	6.0E-05
Pu241	1.440E+01	yr	4.0E-03
Pu242	3.758E+05	yr	9.6E-09
Pu244	8.260E+07	yr	1.0E-15
Am241	4.322E+02	yr	1.2E-04
Am242m	1.520E+02	yr	1.9E-09
Am243	7.380E+03	yr	1.3E-08
Cm243	2.850E+01	yr	1.8E-10
Cm244	1.811E+01	yr	1.0E-07
Cm245	8.500E+03	yr	3.2E-12
Cm246	4.750E+03	yr	7.1E-14
Cm247	1.560E+07	yr	2.5E-20
Cm248	3.390E+05	yr	7.8E-21
Cm250	6.900E+03	yr	2.2E-29
Cf249	3.506E+02	yr	1.7E-20
Cf250	1.308E+01	yr	1.4E-20
Cf251	9.000E+02	yr	3.9E-23
CF252	2.638E_01	yr	1.2E-22

References:

Rhodes, D. W., 1972, letter to Distribution, "Composition of First- and Second-Cycle Wastes", Rhod-4-72.

Schindler, R. E., 1988, *Evaluation of PEW Sources During Fuel Reprocessing Campaign 40*, WIN-257.

Wenzel, D. R., 1977, *Evaluation of Contaminated Soil Site CPP-31 Activity*, EDF Serial Number CPP-058, Functional File Number 1000-50, Revised June 19, 1997.

D. R. Wenzel

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Engineering Design File

Assessment of Radioactivity in INTEC Soil Contamination Site CPP-28

Idaho National Engineering and Environmental Laboratory

Form 412.14
10/9/2003
Rev. 05

EDF No.: 5318 EDF Rev. No.: 0 Project File No.: 23512

1. Title: Assessment of Radioactivity in INTEC Soil Contamination Site CPP-28

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3. NPH Performance Category: _____ or ☒ N/A

4. EDF Safety Category: _____ or ☒ N/A SCC Safety Category: _____ or ☒ N/A

5. Summary:
   This EDF characterizes the radioactivity released to the INTEC tank farm soil in Site CPP-28. This EDF was prepared by DR Wenzel Consulting at BBWI's request.

6. Review (R) and Approval (A) and Acceptance (Ac) Signatures:
   (See instructions for definitions of terms and significance of signatures.)

	R/A	Typed Name/Organization	Signature	Date
Performer/ Author	N/A	D. R. Wenzel/ D. R. Wenzel Consulting		
Performer/ Author	N/A	L. S. Cahn/ Tank Farm Soil Tech Leader		
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Technical Checker	R	M. C. Swenson/ Tank Farm Soils Chemical Eng		
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    N/A.

Assessment of Radioactivity in INTEC Soil Contamination Site CPP-28

1. INTRODUCTION

In October of 1974, a leak of radioactive liquid waste was discovered in the Idaho Nuclear Technology and Engineering Center (INTEC) tank farm (Rigstad 1975). This leak was found to be due to a 1/8-in. drill hole that had been inadvertently drilled into the side of a pipeline used to transfer high-level-radioactive, liquid-waste solutions from the recovery process to the underground liquid waste storage tanks in the tank farm area. Only part of the contaminated soil was removed following discovery of the leak. The remaining contaminated soil is known as contamination site CPP-28 (DOE-ID 2004). The purpose of this Engineering Design File (EDF) is to characterize the radioactivity that was present in the soil at the leak site at the time the leak was investigated in early 1975 to provide a source term that can be used for studies on the transport of the residual activity in the soil.

The best information currently available on the radioactivity in the contaminated soil removed from the site is from the report of the investigation committee (Rigstad 1975). The typical radionuclide spectrum and percentage distribution were determined by sampling the contents of several containers containing contaminated soil removed from the area. The results reported are presented in Table 1. The analysis at the time of the incident investigation did not contain many of the radionuclides of current interest. The relative activities of other radionuclides of interest are given in the subsequent calculations.

Table 1. Measured radionuclide distribution.

Nuclide	%
Mn-52	0.2
Co-60	0.5
Sr, Y-90	59.0
Ru, Rh-106	3.2
Cs-134	1.4
Cs-137	12.2
Ce, Pr-144	21.4
Eu-154	1.3
Eu-155	0.8
Total	1.00

The nomenclature used in the incident report was to combine the activities of the parent and daughter radionuclides. Thus 59% Sr, Y-90 is actually 29.5% Sr-90 and 29.5% Y-90, etc.

While the pipeline was originally installed in the 1955-56 time period, the activity distribution on Table 1 suggests that the leak actually occurred within a year or two prior to discovery of the leak in 1975. The location of the hole in the pipeline was such that leakage did not occur until the pipeline reached a 50%-full level, a condition achieved only when workers apparently neglected to open pertinent

block valves downstream from the hole at the time a liquid waste transfer was initiated though this pipeline. The pipeline was used to transfer high-level radioactive waste from the first-cycle raffinate (IAR) stream to the tank farm.

When fuel was processed at INTEC, the burnup and decay times for individual elements varied. Subsequent calculations are therefore based on pseudo fuel elements, which represent an average of the burnup and decay time for the different fuels processed during an individual campaign.

2. CALCULATIONS

The ORIGEN2 computer code (Croff 1980) was used to calculate the radionuclide distributions as a function of decay time for the types of fuel reprocessed at INTEC prior to discovery of the leak. The first assessment was made for wastes from the reprocessing of aluminum-clad fuels. The dominant aluminum-clad fuels reprocessed at the time of interest were from the Engineering Test Reactor (ETR) and the initial spent fuel from the Advanced Test Reactor (ATR). Fuel from the ETR was selected as the typical aluminum-clad fuel reprocessed before discovery of the leak.

A typical ETR core cycle operated at an average of 164.9 MW for 650 h (IDO 1960). ETR contained 51 elements with 500 g U-235 and 16 elements at 230 g U-235. Thus the core contained 29.18 kg of U-235. The fuel enrichment was 93.3% U-235. This calculated to be 5.651 MW/kg U-235. Calculating an inventory for a 500-g element, the average power level was 2.826 MW for a period of 650 h.

The input and summarized output for ORIGEN2 run ETR2 are presented in Appendix A. The calculated radionuclide activities as a function of decay time for the radionuclides used to evaluate the inventory for aluminum-clad fuel are presented in Table 2. The ratio of the Cs isotopes Cs-134 and Cs-137 was used to determine the age of the waste. These isotopes are used because of their vastly different half-lives, 2.062 years for Cs-134 and 30.17 years for Cs-137. Another reason for choosing these Cs isotopes is that being of the same element, their ratio is not affected by the soil absorption rate. A decay time of 1.9 years, shown by the gray shading in Table 2, yielded the best fit for the measured Cs-134/Cs-137 ratio of 0.115.

Table 3 presents ratios of the various radionuclides as a function of decay time. The last column in Table 3 divides the radionuclide ratios from the ORIGEN2-calculated inventory by the ratios determined from the sample results. The column for a decay time of 1.9 years was used for comparison because the

Table 2. Radionuclide activities used to evaluate aluminum-clad fuel for the leak.

	Aluminum-Clad Fuel - ETR 500-g Element Activity (Ci)							
Radionuclide	1.6 Year	1.7 Year	1.8 Year	1.9 Year	2.0 Year	2.1 Year	2.2 Year	2.3 Year
Sr-90	2.30E+02	2.29E+02	2.29E+02	2.28E+02	2.28E+02	2.27E+02	2.26E+02	2.26E+02
Ru-106	1.65E+02	1.54E+02	1.44E+02	1.35E+02	1.26E+02	1.17E+02	1.10E+02	1.02E+02
Cs-134	3.01E+01	2.91E+01	2.81E+01	2.72E+01	2.63E+01	2.54E+01	2.46E+01	2.38E+01
Cs-137	2.39E+02	2.38E+02	2.38E+02	2.37E+02	2.37E+02	2.36E+02	2.35E+02	2.35E+02
Ce-144	1.97E+03	1.80E+03	1.65E+03	1.51E+03	1.38E+03	1.26E+03	1.15E+03	1.06E+03
Eu-154	2.46E+00	2.44E+00	2.42E+00	2.40E+00	2.39E+00	2.37E+00	2.35E+00	2.33E+00
Eu-155	4.34E+00	4.28E+00	4.22E+00	4.16E+00	4.10E+00	4.05E+00	3.99E+00	3.94E+00

Table 3. Comparison of radionuclide ratios from aluminum-clad fuel to measured soil activity.

	ETR aluminum-clad fuel									
Nuclide Ratios	1.6 Year	1.7 Year	1.8 Year	1.9 Year	2.0 Year	2.1 Year	2.2 Year	2.3 Year	Soil	1.9 Year Al/Soil
Sr-90/Ru-106	1.39E+00	1.48E+00	1.59E+00	1.70E+00	1.81E+00	1.94E+00	2.07E+00	2.21E+00	1.84E+01	9.20E-02
Sr-90/Cs-134	7.64E+00	7.89E+00	8.14E+00	8.40E+00	8.66E+00	8.94E+00	9.22E+00	9.51E+00	2.11E+01	3.99E-01
Sr-90/Cs-137	9.62E-01	9.62E-01	9.62E-01	9.62E-01	9.62E-01	9.62E-01	9.62E-01	9.62E-01	2.42E+00	3.98E-01
Sr-90/Ce-144	1.17E-01	1.27E-01	1.39E-01	1.51E-01	1.65E-01	1.80E-01	1.96E-01	2.14E-01	2.76E+00	5.49E-02
Sr-90/Eu-154	9.33E+01	9.37E+01	9.43E+01	9.49E+01	9.54E+01	9.59E+01	9.65E+01	9.70E+01	2.27E+01	4.18E+00
Sr-90/Eu-155	5.29E+01	5.35E+01	5.42E+01	5.48E+01	5.54E+01	5.61E+01	5.67E+01	5.74E+01	3.69E+01	1.49E+00
Ru-106/Cs-134	5.50E+00	5.31E+00	5.13E+00	4.95E+00	4.78E+00	4.62E+00	4.46E+00	4.30E+00	1.14E+00	4.33E+00
Ru-106/Cs-137	6.93E-01	6.48E-01	6.06E-01	5.67E-01	5.31E-01	4.97E-01	4.65E-01	4.35E-01	1.31E-01	4.33E+00
Ru-106/Ce-144	8.40E-02	8.58E-02	8.75E-02	8.93E-02	9.11E-02	9.30E-02	9.50E-02	9.69E-02	1.50E-01	5.97E-01
Ru-106/Eu-154	6.72E+01	6.32E+01	5.94E+01	5.59E+01	5.27E+01	4.96E+01	4.67E+01	4.39E+01	1.23E+00	4.55E+01
Ru-106/Eu-155	3.81E+01	3.61E+01	3.41E+01	3.23E+01	3.06E+01	2.90E+01	2.74E+01	2.60E+01	2.00E+00	1.62E+01
Cs-134/Cs-137	1.26E-01	1.22E-01	1.18E-01	1.15E-01	1.11E-01	1.08E-01	1.04E-01	1.01E-01	1.15E-01	9.98E-01
Cs-134/Ce-144	1.53E-02	1.61E-02	1.71E-02	1.80E-02	1.91E-02	2.01E-02	2.13E-02	2.25E-02	1.31E-01	1.38E-01
Cs-134/Eu-154	1.22E+01	1.19E+01	1.16E+01	1.13E+01	1.10E+01	1.07E+01	1.05E+01	1.02E+01	1.08E+00	1.05E+01
Cs-134/Eu-155	6.92E+00	6.79E+00	6.65E+00	6.53E+00	6.40E+00	6.28E+00	6.15E+00	6.03E+00	1.75E+00	3.73E+00
Cs-137/Ce-144	1.21E-01	1.32E-01	1.44E-01	1.57E-01	1.72E-01	1.87E-01	2.04E-01	2.23E-01	1.14E+00	1.38E-01
Cs-137/Eu-154	9.69E+01	9.75E+01	9.80E+01	9.86E+01	9.92E+01	9.97E+01	1.00E+02	1.01E+02	9.38E+00	1.05E+01
Cs-137/Eu-155	5.50E+01	5.57E+01	5.63E+01	5.70E+01	5.76E+01	5.83E+01	5.90E+01	5.97E+01	1.53E+01	3.74E+00
Ce-144/Eu-154	7.99E+02	7.36E+02	6.79E+02	6.27E+02	5.78E+02	5.33E+02	4.91E+02	4.53E+02	8.23E+00	7.62E+01
Ce-144/Eu-155	4.53E+02	4.21E+02	3.90E+02	3.62E+02	3.36E+02	3.12E+02	2.89E+02	2.68E+02	1.34E+01	2.71E+01
Eu-154/Eu-155	5.68E-01	5.71E-01	5.74E-01	5.78E-01	5.81E-01	5.85E-01	5.88E-01	5.91E-01	1.63E+00	3.55E-01

calculated ratio of Cs-134/Cs-137 divided by the measured ratio is very close to 1. However, none of the other calculated ratios divided by the measured ratios come very close to 1. The conclusion reached is that the measured activity does not have a fingerprint of only aluminum-clad fuel reprocessing by itself.

Another ORIGEN2 run was made, this time for generic zirconium-clad fuel (see Table 4). The ORIGEN2 run was based on a previous calculation used to evaluate the radionuclide inventory in Zr calcine (Wenzel 1997). The input and summarized output for ORIGEN2 run ZR2 are presented in Appendix B. Table 5 presents ratios of the various radionuclides as a function of decay time for generic zirconium-clad fuel. As in Table 3, the last column in Table 5 divides the radionuclides ratios calculated from the ORIGEN2 results by the ratios determined from the sample results. The column for a decay time of 6.9 years was used for comparison because the calculated ratio of Cs-134/Cs-137 divided by the measured ratio is very close to 1. The closer the values in the last column are to 1, the better the data fit. The radionuclide ratios that are approaching 1 are shaded gray. While the date for zirconium-clad fuel is somewhat better than that for aluminum-clad fuel, the fit still is not very good. The conclusion reached is that the measured activity does not represent the reprocessing of zirconium-clad fuel by itself very well.

The next calculations made were to evaluate how well coprocessing wastes fit the radionuclide distribution measured in the contaminated soil. As noted above, the evaluation of the aluminum-clad fuel indicated that the decay age of the aluminum-clad fuel was about 1.9 years at the time the leak was discovered. This places the leak most probably during reprocessing campaign as Run No. 30 which ran between February and May of 1974 (Staiger 2003). The uranium mass ratio for aluminum/zirconium fuel during reprocessing campaign Run No. 30 was approximately 1.5. Table 6 presents the calculated ratios for coprocessing.

The last column in Table 6 presents the radionuclide ratios calculated from the ORIGEN2 results divided by the measured ratios from the soil analyses. As before, the closer the data in the last column is to 1, the better the calculated data fit the measured data. As shown in Table 6, the ratios Ru-106/Cs-134, Ru-106/Cs-137, and Cs-134/Cs-137 (shaded gray) all are very good. Five of the ratios shown as bold in Table 6 are within a factor of 2. Ratios that exceed a factor of 2 all involve Sr or Eu. It is not known if an actual Sr-90 analysis was done or whether all beta activity not associated with the other identified radionuclides was considered to be Sr-90. If the latter method were used, the Sr-90 activity reported would be expected to be high. While the ratio of Eu-154/Eu-155 is quite good, the ratio of Eu to Cs and Ce isotopes is poor. This may be because of the difference in migration rates of these elements in the soil. The conclusion reached is that coprocessing wastes consisting of the reprocessing of 1.9-year-old aluminum-clad fuel and 6.9-year-old zirconium-clad fuel best represents the activity removed from soil site CPP-28 in early 1975.

Table 4. Radionuclide activities used to evaluate zirconium-clad fuel for the leak.

	Zirconium-Clad Fuel - Generic Element Activity (Ci)							
Radionuclide	6.6 Year	6.7 Year	6.8 Year	6.9 Year	7.0 Year	7.1 Year	7.2 Year	7.3 Year
Sr- 90	1.18E+03	1.18E+03	1.18E+03	1.17E+03	1.17E+03	1.17E+03	1.17E+03	1.16E+03
Ru-106	9.29E+00	8.67E+00	8.09E+00	7.55E+00	7.05E+00	6.58E+00	6.15E+00	5.74E+00
Cs-134	1.58E+02	1.53E+02	1.48E+02	1.43E+02	1.38E+02	1.34E+02	1.29E+02	1.25E+02
Cs-137	1.23E+03	1.23E+03	1.23E+03	1.23E+03	1.22E+03	1.22E+03	1.22E+03	1.21E+03
Ce-144	3.20E+01	2.93E+01	2.68E+01	2.45E+01	2.24E+01	2.05E+01	1.88E+01	1.72E+01
Eu-54	4.11E+01	4.07E+01	4.04E+01	4.01E+01	3.98E+01	3.94E+01	3.91E+01	3.88E+01
Eu-155	1.48E+01	1.46E+01	1.44E+01	1.42E+01	1.40E+01	1.38E+01	1.36E+01	1.34E+01

Table 5. Comparison of radionuclide ratios from zirconium-clad fuel to measured soil activity.

	Zirconium-Clad Fuel									
Nuclide Ratios	6.6 Year	6.7 Year	6.8 Year	6.9 Year	7.0 Year	7.1 Year	7.2 Year	7.3 Year	Soil	7.1 Year Zr/Soil
Sr-90/Ru-106	1.27E+02	1.36E+02	1.45E+02	1.55E+02	1.66E+02	1.77E+02	1.90E+02	2.03E+02	1.84E+01	8.42E+00
Sr-90/Cs-134	7.47E+00	7.70E+00	7.95E+00	8.20E+00	8.46E+00	8.73E+00	9.01E+00	9.30E+00	2.11E+01	3.89E-01
Sr-90/Cs-137	9.57E-01	9.57E-01	9.58E-01	9.58E-01	9.57E-01	9.57E-01	9.57E-01	9.57E-01	2.42E+00	3.96E-01
Sr-90/Ce-144	3.69E+01	4.02E+01	4.39E+01	4.78E+01	5.21E+01	5.68E+01	6.20E+01	6.76E+01	2.76E+00	1.73E+01
Sr-90/Eu-154	2.88E+01	2.89E+01	2.91E+01	2.93E+01	2.94E+01	2.96E+01	2.98E+01	2.99E+01	2.27E+01	1.29E+00
Sr-90/Eu-155	8.00E+01	8.10E+01	8.20E+01	8.29E+01	8.38E+01	8.48E+01	8.59E+01	8.68E+01	3.69E+01	2.25E+00
Ru-106/Cs-134	5.87E-02	5.67E-02	5.47E-02	5.28E-02	5.10E-02	4.92E-02	4.75E-02	4.59E-02	1.14E+00	4.62E-02
Ru-106/Cs-137	7.52E-03	7.04E-03	6.59E-03	6.17E-03	5.77E-03	5.40E-03	5.05E-03	4.73E-03	1.31E-01	4.70E-02
Ru-106/Ce-144	2.90E-01	2.96E-01	3.02E-01	3.08E-01	3.14E-01	3.21E-01	3.27E-01	3.34E-01	1.50E-01	2.06E+00
Ru-106/Eu-154	2.26E-01	2.13E-01	2.00E-01	1.89E-01	1.77E-01	1.67E-01	1.57E-01	1.48E-01	1.23E+00	1.53E-01
Ru-106/Eu-155	6.29E-01	5.96E-01	5.64E-01	5.34E-01	5.05E-01	4.78E-01	4.53E-01	4.29E-01	2.00E+00	2.67E-01
Cs-134/Cs-137	1.28E-01	1.24E-01	1.20E-01	1.17E-01	1.13E-01	1.10E-01	1.06E-01	1.03E-01	1.15E-01	1.02E+00
Cs-134/Ce-144	4.94E+00	5.22E+00	5.52E+00	5.83E+00	6.16E+00	6.51E+00	6.88E+00	7.28E+00	1.31E-01	4.46E+01
Cs-134/Eu-154	3.85E+00	3.76E+00	3.66E+00	3.57E+00	3.48E+00	3.39E+00	3.31E+00	3.22E+00	1.08E+00	3.31E+00
Cs-134/Eu-155	1.07E+01	1.05E+01	1.03E+01	1.01E+01	9.91E+00	9.72E+00	9.53E+00	9.34E+00	1.75E+00	5.77E+00
Cs-137/Ce-144	3.85E+01	4.20E+01	4.58E+01	4.99E+01	5.45E+01	5.94E+01	6.48E+01	7.07E+01	1.14E+00	4.38E+01
Cs-137/Eu-154	3.01E+01	3.02E+01	3.04E+01	3.06E+01	3.07E+01	3.09E+01	3.11E+01	3.13E+01	9.38E+00	3.26E+00
Cs-137/Eu-155	8.36E+01	8.46E+01	8.56E+01	8.66E+01	8.75E+01	8.87E+01	8.97E+01	9.07E+01	1.53E+01	5.68E+00
Ce-144/Eu-154	7.81E-01	7.20E-01	6.64E-01	6.12E-01	5.65E-01	5.21E-01	4.80E-01	4.43E-01	8.23E+00	7.44E-02
Ce-144/Eu-155	2.17E+00	2.01E+00	1.87E+00	1.73E+00	1.61E+00	1.49E+00	1.38E+00	1.28E+00	1.34E+01	1.30E-01
Eu-154/Eu-155	2.78E+00	2.80E+00	2.82E+00	2.83E+00	2.85E+00	2.87E+00	2.88E+00	2.90E+00	1.63E+00	1.74E+00

Table 6. Radionuclide activities used to evaluate coprocessing waste for the leak.

	Coprocessing									
			Al * 1.5	Zr * 1	Al age = 1.9 Year		Zr age = 6.9 Year			
Nuclide Ratios	6.6 Year	6.7 Year	6.8 Year	6.9 Year	7.0 Year	7.1 Year	7.2 Year	7.3 Year	Soil	Co/Soil
Sr-90/Ru-106	7.22E+00	7.22E+00	7.23E+00	7.24E+00	7.24E+00	7.24E+00	7.25E+00	7.25E+00	1.84E+01	3.93E-01
Sr-90/Cs-134	7.66E+00	7.85E+00	8.05E+00	8.25E+00	8.45E+00	8.65E+00	8.86E+00	9.08E+00	2.11E+01	3.91E-01
Sr-90/Cs-137	9.58E-01	9.58E-01	9.59E-01	9.59E-01	9.58E-01	9.58E-01	9.58E-01	9.58E-01	2.42E+00	3.96E-01
Sr-90/Ce-144	6.64E-01	6.64E-01	6.64E-01	6.63E-01	6.62E-01	6.62E-01	6.61E-01	6.60E-01	2.76E+00	2.41E-01
Sr-90/Eu-154	3.41E+01	3.43E+01	3.45E+01	3.47E+01	3.49E+01	3.51E+01	3.53E+01	3.55E+01	2.27E+01	1.53E+00
Sr-90/Eu-155	7.25E+01	7.31E+01	7.37E+01	7.43E+01	7.48E+01	7.54E+01	7.61E+01	7.67E+01	3.69E+01	2.01E+00
Ru-106/Cs-134	1.06E+00	1.09E+00	1.11E+00	1.14E+00	1.17E+00	1.19E+00	1.22E+00	1.25E+00	1.14E+00	9.97E-01
Ru-106/Cs-137	1.33E-01	1.33E-01	1.33E-01	1.32E-01	1.32E-01	1.32E-01	1.32E-01	1.32E-01	1.31E-01	1.01E+00
Ru-106/Ce-144	9.21E-02	9.19E-02	9.17E-02	9.16E-02	9.15E-02	9.13E-02	9.12E-02	9.11E-02	1.50E-01	6.13E-01
Ru-106/Eu-154	4.73E+00	4.75E+00	4.77E+00	4.79E+00	4.82E+00	4.84E+00	4.87E+00	4.89E+00	1.23E+00	3.89E+00
Ru-106/Eu-155	1.00E+01	1.01E+01	1.02E+01	1.03E+01	1.03E+01	1.04E+01	1.05E+01	1.06E+01	2.00E+00	5.13E+00
Cs-134/Cs-137	1.25E-01	1.22E-01	1.19E-01	1.16E-01	1.13E-01	1.11E-01	1.08E-01	1.06E-01	1.15E-01	1.01E+00
Cs-134/Ce-144	8.68E-02	8.46E-02	8.25E-02	8.04E-02	7.84E-02	7.65E-02	7.46E-02	7.28E-02	1.31E-01	6.15E-01
Cs-134/Eu-154	4.45E+00	4.37E+00	4.29E+00	4.21E+00	4.13E+00	4.05E+00	3.98E+00	3.91E+00	1.08E+00	3.91E+00
Cs-134/Eu-155	9.47E+00	9.32E+00	9.16E+00	9.01E+00	8.86E+00	8.72E+00	8.58E+00	8.45E+00	1.75E+00	5.15E+00
Cs-137/Ce-144	6.93E-01	6.93E-01	6.92E-01	6.92E-01	6.91E-01	6.91E-01	6.90E-01	6.89E-01	1.14E+00	6.07E-01
Cs-137/Eu-154	3.56E+01	3.58E+01	3.60E+01	3.62E+01	3.64E+01	3.66E+01	3.68E+01	3.70E+01	9.38E+00	3.86E+00
Cs-137/Eu-155	7.57E+01	7.63E+01	7.69E+01	7.75E+01	7.81E+01	7.88E+01	7.94E+01	8.00E+01	1.53E+01	5.08E+00
Ce-144/Eu-154	5.13E+01	5.17E+01	5.20E+01	5.23E+01	5.27E+01	5.30E+01	5.33E+01	5.37E+01	8.23E+00	6.36E+00
Ce-144/Eu-155	1.09E+02	1.10E+02	1.11E+02	1.12E+02	1.13E+02	1.14E+02	1.15E+02	1.16E+02	1.34E+01	8.38E+00
Eu-154/Eu-155	2.13E+00	2.13E+00	2.14E+00	2.14E+00	2.15E+00	2.15E+00	2.16E+00	2.16E+00	1.63E+00	1.32E+00

Adjustments to the calculated source term are necessary for H-3 and I-129, both of which are volatile. Based on an I-129 behavior study (McManus 1982) for INTEC fuel reprocessing, the IAR contains 85% of the I-129 present in the first-cycle solvent extraction.

The amount of H-3 present was established from the analysis of liquid samples in 1971 of deep tank WM-185 which contained IAR waste from coprocessing (Rhodes 1972). The ratio of H-3/Cs-137 in deep tank WM-185 was used as the most likely ratio of H-3/Cs-137 in the leak of interest. The measured H-3/Cs-137 ratio of 8.14E-04 in WM-185 has been used to estimate the amount of H-3 present in the leaking IAR solution.

The purpose of reprocessing was to remove the U from the spent fuel. The amount of U calculated using the ORIGEN2 code (see Appendixes A and B) is for the spent fuel, not the IAR stream. The chemical analysis of the 1971 sample from deep tank WM-185 for U (Rhodes 1972) was used to determine an activity ratio of U-235/Cs-137 of 5.24E-10 calculated as follows:

$$\left[4.7E{-}04\frac{g}{L}U\right]\left[.516\frac{U-235}{U}\right]\left[2.161E{-}06\frac{Ci}{g\,U{-}235}\right]\left[\frac{L}{10^3\,mL}\right]\left[3.7E{+}10\,\frac{d/s}{Ci}\right]=1.94E{-}02\frac{d/s}{mL}U-235$$

$$\frac{1.94E{-}02\dfrac{d/s}{mL}U{-}235}{3.7E{+}07\dfrac{d/s}{mL}Cs{-}137}=5.24E{-}10$$

During this same time period, there was also an Np-237 recovery process being operated. The result of analysis of the 1971 sample from WM-185 was 1.19E-5 g/L Np (Rhodes 1972). It was assumed that all of the Np was Np-237. A ratio of Np-237/Cs-137 of 8.38E-9 was calculated as follows:

$$\left[1.19E{-}05\frac{g}{L}Np{-}237\right]\left[7.049E{-}04\frac{Ci}{g\,Np{-}237}\right]\left[\frac{L}{10^3\,mL}\right]\left[3.7E{+}10\,\frac{d/s}{Ci}\right]=3.10E{-}01\frac{d/s}{mL}Np{-}237$$

$$\frac{3.10E{-}01\dfrac{d/s}{mL}Np{-}237}{3.7E{+}07\dfrac{d/s}{mL}Cs{-}137}=8.38E{-}9$$

A coprocessing waste source term for the leak was established by summing the radionuclide inventory in Appendix A for aluminum-clad fuel, weighted by a factor of 1.5, and the inventory in Appendix B for zirconium-clad fuel, weighted by a factor of 1. Table 7 contains the calculated source term for the leak, normalized to an activity of 1 for Cs-137. The radionuclide activities presented in Table 7 are the activities that were present in early 1975, the time that they were measured for the incident report. The output from the ORIGEN2 calculations contains far more radionuclides than are of interest for soil migration studies. All radionuclides with half-lives less than 1 year have been deleted and the results have been rounded to two places of accuracy. Radionuclides, which have inventories of less than one atom as calculated by ORIGEN2, have also been deleted from the table.

Table 7. Calculated radionuclide activities for Site CPP-28.

Nuclide	Half-life	Activity Normalized to Cs-137
H-3	1.233E+01 yr	8.1E-04
Be-10	1.510E+06 yr	3.3E-11
C-14	5.730E+03 yr	1.3E-09
Se-79	1.130E+06 yr	4.8E-06
Rb-87	4.750E+10 yr	3.2E-10
Sr-90	2.878E+01 yr	9.6E-01
Zr-93	1.530E+06 yr	2.5E-05
Nb-93m	1.613E+01 yr	7.6E-06
Nb-94	2.030E+04 yr	2.5E-10
Tc-98	4.200E+06 yr	2.6E-11
Tc-99	2.111E+05 yr	1.6E-04
Ru-106	1.023E+00 yr	1.3E-01
Pd-107	6.500E+06 yr	1.7E-07
Ag-108m	4.180E+02 yr	5.2E-12
Cd-113m	1.410E+01 yr	1.1E-04
In-115	4.410E+14 yr	7.6E-17
Sn-121m	5.500E+01 yr	9.3E-07
Te-123	1.000E+13 yr	4.7E-18
Sb-125	2.758E+00 yr	1.7E-02
Sn-126	1.000E+05 yr	4.2E-06
I-129	1.570E+07 yr	2.2E-07
Cs-134	2.065E+00 yr	1.2E-01
Cs-135	2.300E+06 yr	6.9E-06
Cs-137	3.007E+01 yr	1.0E+00
La-138	1.050E+11 yr	2.1E-15
Nd-144	2.290E+15 yr	1.7E-14
Pm-146	5.530E+00 yr	6.5E-06
Sm-146	1.030E+08 yr	2.1E-12
Pm-147	2.623E+00 yr	7.6E-01
Sm-147	1.060E+11 yr	6.2E-11
Sm-148	7.000E+15 yr	4.6E-16
Sm-149	2.000E+15 yr	2.7E-17
Eu-150	3.690E+01 yr	1.7E-10
Sm-151	9.000E+01 yr	4.6E-03
Eu-152	1.354E+01 yr	9.6E-05
Gd-152	1.080E+14 yr	1.7E-17
Eu-154	8.593E+00 yr	2.8E-02
Eu-155	4.761E+00 yr	1.3E-02
Ho-166m	1.200E+03 yr	5.4E-10
Tm-171	1.920E+00 yr	3.3E-12

Table 7. (continued).

Nuclide	Half-life	Activity Normalized to Cs-137
Pb-210	2.230E+01 yr	1.0E-14
Bi-208	3.680E+05 yr	3.6E-24
Bi-210m	3.040E+06 yr	3.1E-24
Ra-226	1.600E+03 yr	2.6E-13
Ra-228	5.750E+00 yr	7.3E-16
Ac-227	2.177E+01 yr	2.4E-11
Th-228	1.912E+00 yr	9.6E-08
Th-229	7.340E+03 yr	5.3E-13
Th-230	7.538E+04 yr	5.7E-10
Th-232	1.405E+10 yr	2.1E-15
Pa-231	3.276E+04 yr	1.7E-10
U-232	6.890E+01 yr	4.5E-10
U-233	1.592E+05 yr	9.8E-14
U-234	2.455E+05 yr	1.2E-08
U-235	7.038E+08 yr	5.2E-10
U-236	2.342E+07 yr	2.0E-09
U-238	4.470E+09 yr	4.1E-12
Np-237	2.144E+06 yr	8.4E-09
Pu-236	2.858E+00 yr	4.7E-07
Pu-238	8.770E+01 yr	1.2E-02
Pu-239	2.411E+04 yr	4.3E-05
Pu-240	6.563E+03 yr	1.5E-05
Pu-241	1.435E+01 yr	1.7E-03
Pu-242	3.733E+05 yr	4.9E-09
Pu-244	8.080E+07 yr	3.1E-16
Am-241	4.322E+02 yr	1.0E-05
Am-242m	1.410E+02 yr	6.1E-08
Am-243	7.370E+03 yr	1.7E-08
Cm-243	2.910E+01 yr	1.6E-08
Cm-244	1.810E+01 yr	6.8E-07
Cm-245	8.500E+03 yr	4.1E-11
Cm-246	4.730E+03 yr	4.3E-12
Cm-247	1.560E+07 yr	6.8E-18
Cm-248	3.400E+05 yr	9.3E-18
Cm-250	9.000E+03 yr	1.9E-25
Cf-249	3.510E+02 yr	6.2E-17
Cf-250	1.308E+01 yr	1.6E-16
Cf-251	8.980E+02 yr	1.1E-18
Cf-252	2.645E+00 yr	1.8E-17
Total		3.0

Radiation levels at the leak site were measured in 1975 by driving 20-ft lengths of standard 2-in., Schedule 80, carbon-steel pipe equipped with a formed drill point at one end (Rigstad 1975). These pipes were driven into the ground and a radiation detector probe was lowered into the test-pipe, allowing measurement of the direct radiation field as a function of depth. A copy of the measured radiation profile is presented in Appendix C.

The direct radiation field from the radionuclide distribution presented in Table 7 in soil has been calculated using the MicroShield Computer Code (Grove Engineering 2003). The source geometry was modeled as an annular cylinder with a source thickness of 1 ft and a height of 3 ft. The inside of the annular cylinder represents the 2-in., Schedule 80, carbon-steel pipe from which direct radiation measurements were taken. This pipe has an internal diameter of 1.929 in. and a thickness of 0.218 in. This calculates to be an internal radius of 2.45 cm and a thickness of 0.554 cm. The radionuclide concentration was assumed to be uniform over the source volume. Using the above dimensions, the calculated radiation field is within 97% of that from a source of infinite dimensions. The soil density used in the MicroShield calculation was assumed to be 2 g/cm^3, the same as assumed by the investigating committee in their calculations. Since MicroShield does not have soil in its library of materials, concrete was used as the characteristic material for soil. The MicroShield run for this calculation is presented in Appendix D. All of the radionuclides in Appendix A and Appendix B were used in the shielding calculation. The calculated radiation field of 2.53E+03 mR/h is for the source volume containing 1 Ci of Cs-137 and all of the other associated radionuclides.

By dividing the calculated radiation field (2.38E+03) by the source volume used in the MicroShield run (3.20E+05 cm^3), a conversion factor can be calculated which can be used to estimate the total radioactivity that was present in the soil at the time that the direct radiation measurements were made. A Cs-137 activity of 1 Ci / 3.20E+05 cm^3 yields a concentration of 3.13E-6 Ci Cs-137 / cm^3. A conversion factor is calculated as:

$$conversion\ factor = \frac{3.13E-06\ Ci\ Cs-137/cm^3}{2.38E+03\ mR/h} = 1.32E-09\ \frac{Ci\ Cs-137/cm^3}{mR/h}$$

The concentrations of all other radionuclides are calculated by multiplying the Ci of Cs-137 calculated using the above conversion factor by the normalized activities of each of the radionuclides as presented in Table 7.

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